

LA-UR-21-29850

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Title: Resolving the Spinon Continuum

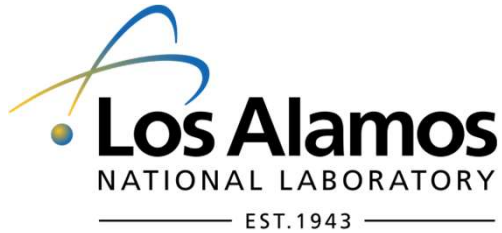
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Intended for: Report

Issued: 2021-10-04

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Resolving the Spinon Continuum

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Los Alamos National Laboratory

Institute of Material Science Rapid Response Roundup
October 5, 2021

LA-UR-##-####

Classifying States of Matter

Landau Theory: Spontaneously Broken Symmetry

$$F(T) = F_0(T) + \alpha_0(T - T_c)\eta^2 + \frac{1}{2}\beta\eta^4$$

$$\frac{dF}{d\eta} = 0 = 2\alpha_0(T - T_c)\eta + 2\beta\eta^3$$

$$T > T_c$$

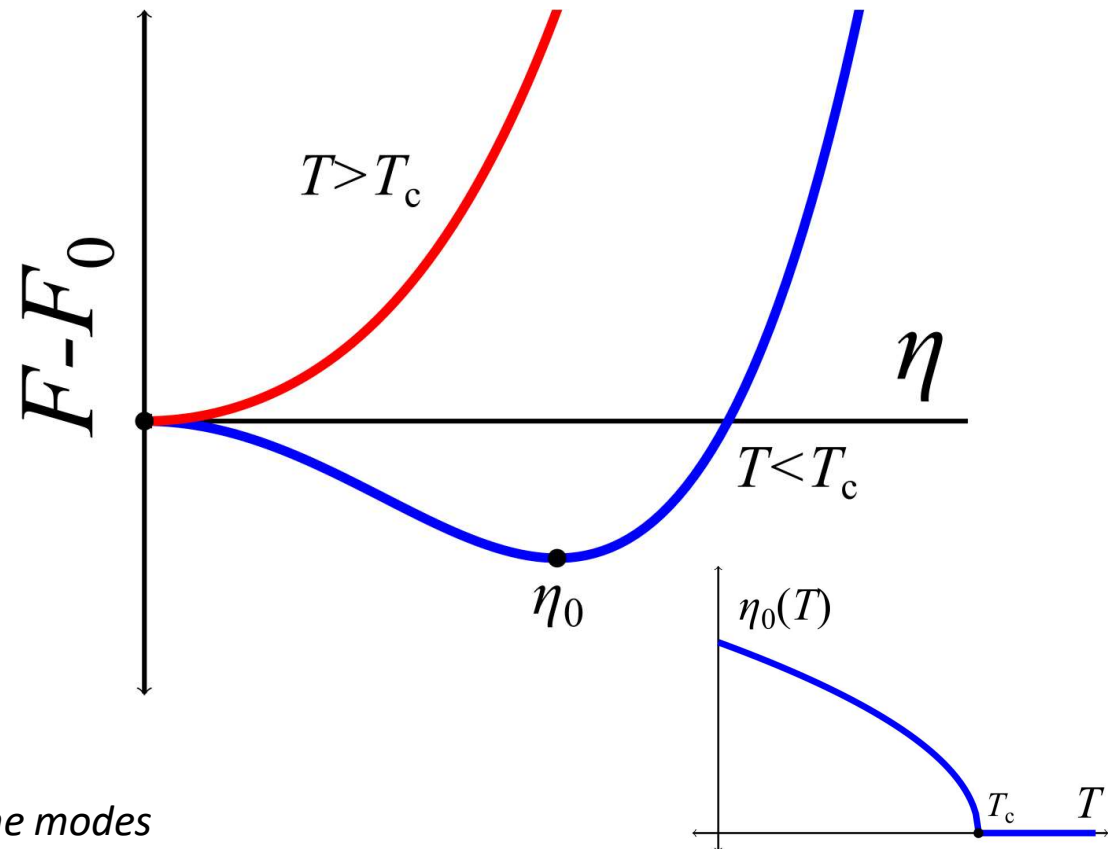
$$\eta = 0$$

$$T < T_c$$

$$\eta = \pm \sqrt{\alpha_0/\beta (T_c - T)}$$

Ordered state of matter:

- Emergent OP
- Entropy release $\Delta S < 0$
- Long wavelength fluctuations in the OP = *Goldstone modes*



Quantum Spin Liquids

Topological phases of matter follow a different classification scheme rooted in the geometric character of the electronic wave function

- *Symmetry (and symmetry breaking) still plays an important role in protecting topological invariants*

Spin liquids are quantum disordered states in $D > 1$

- No universally accepted experimental evidence for the existence of a spin liquid ground state in any one material

Fundamental characteristics:

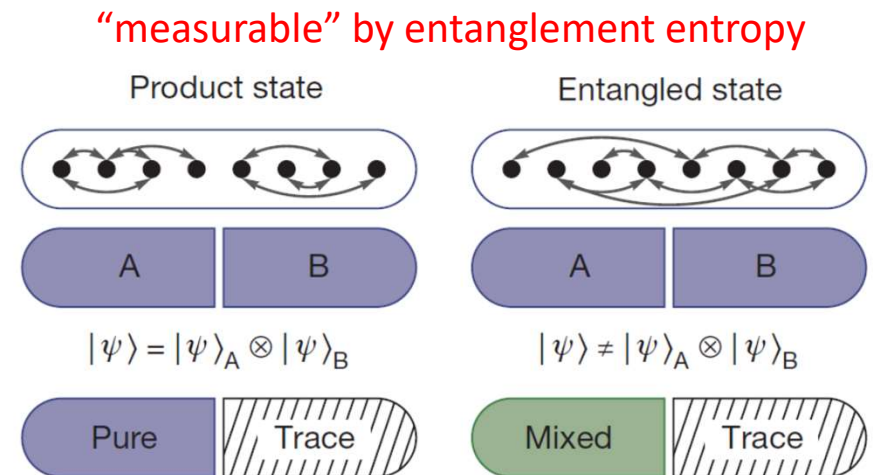
- 1) Absence of long range magnetic order $T \rightarrow 0$
- 2) Long range quantum entanglement in ground state
- 3) Exotic “fractional” excitations

Gapped Z_2 QSL excitations fractionalizes into:

Spinons – Gauge charge

Visons – Gauge flux

Can be probed spectroscopically!



Nature **528**, 77 (2015)

Fractional Excitations: Spinons

$AFM_1 = \uparrow \downarrow \uparrow \downarrow \uparrow \downarrow \uparrow \downarrow \uparrow \downarrow$

$AFM_2 = \downarrow \uparrow \downarrow \uparrow \downarrow \uparrow \downarrow \uparrow \downarrow \uparrow$

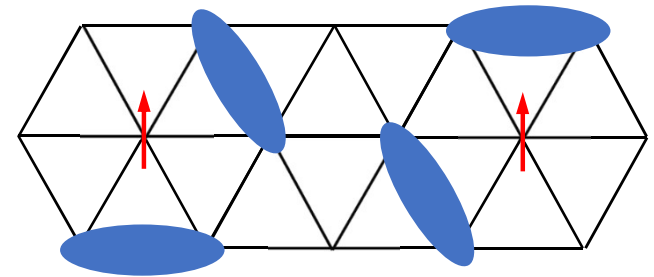
$\Delta S = 1 \rightarrow 2 \times \text{Domain Wall}$

$\uparrow \downarrow \uparrow \downarrow \downarrow \uparrow \downarrow \uparrow \downarrow \uparrow \downarrow$

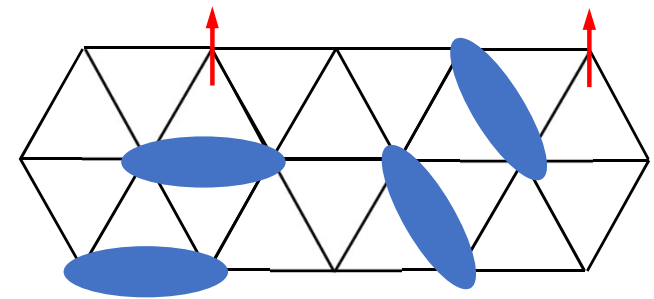
$AFM_1 \quad AFM_2 \quad AFM_1$

$\uparrow \downarrow \downarrow \uparrow \downarrow \uparrow \downarrow \uparrow \downarrow \uparrow \downarrow$

1D: AFM is quantum disordered GS
Spinon = Propagating domain wall

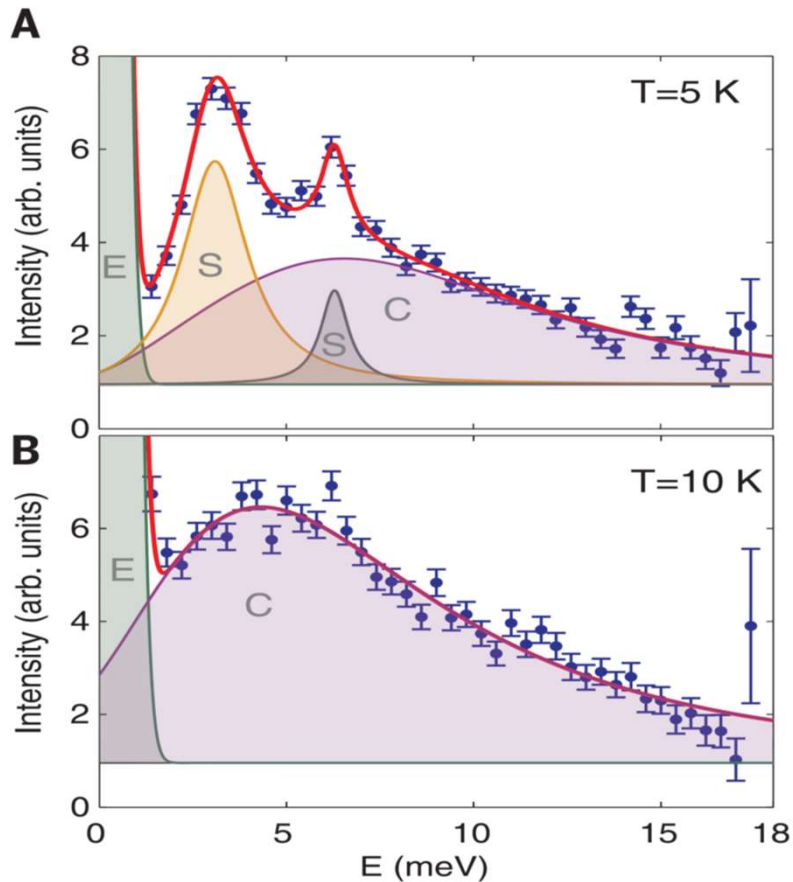


$$\text{blue oval} = \frac{1}{\sqrt{2}} (|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)$$



2D: Spinon can propagate with little energy cost in a RVB state \sim asymptotic freedom in QCD

Trouble with Probing the Linear Response



Science **356**, 1055 (2017)

Deconfined spinons can take on a continuous energy distribution resulting in a broad excitation (> 10 meV) which constitutes all possible ways energy and momentum are shared amongst fractionalized QP

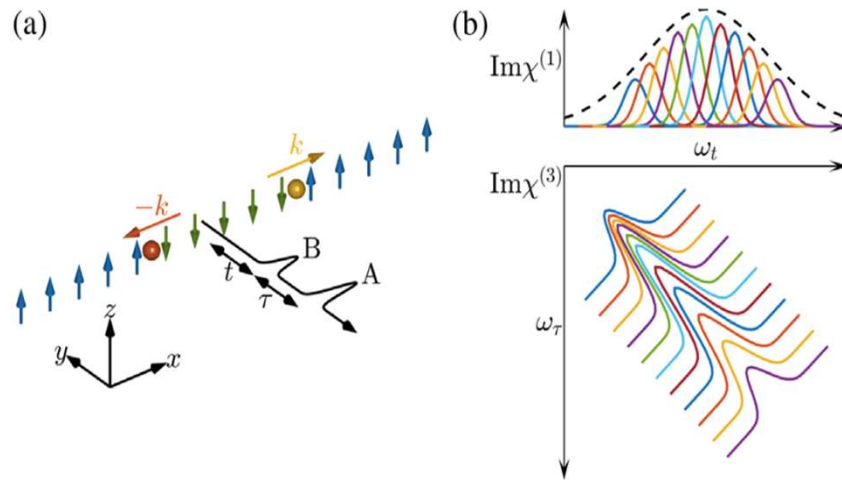
Linear probes (INS, IXS, Raman) can only probe an envelope of excitations leading to ambiguity in the spectrum.

- Anharmonic spin wave excitation
- multimagnon excitation
- spin wave dampening
- quasi-particle “incoherence” (magnon breakdown)
- Disorder

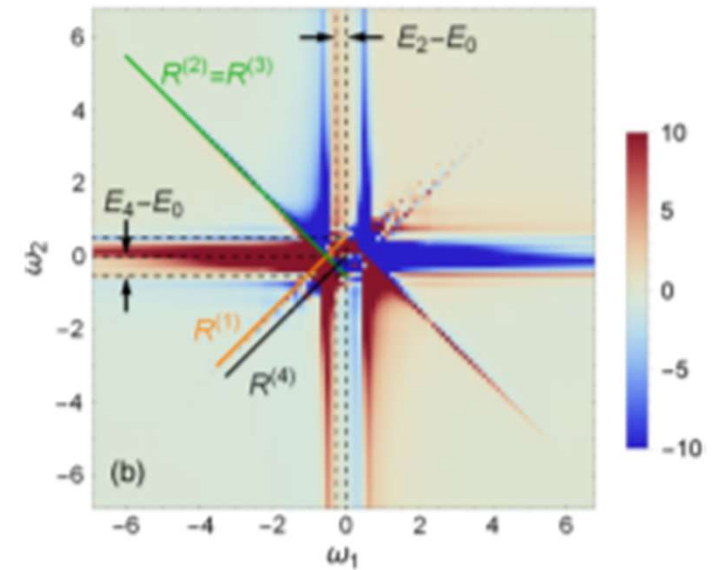
- *Nature Comm.* **8**, 1152 (2017)

A clear signature of fractionalized excitations necessitates that the spinon continuum be resolved

Using 2D Coherent Spectroscopy to Resolve the Spinon Continuum



Phys. Rev. Lett. **122**, 257401 (2019)



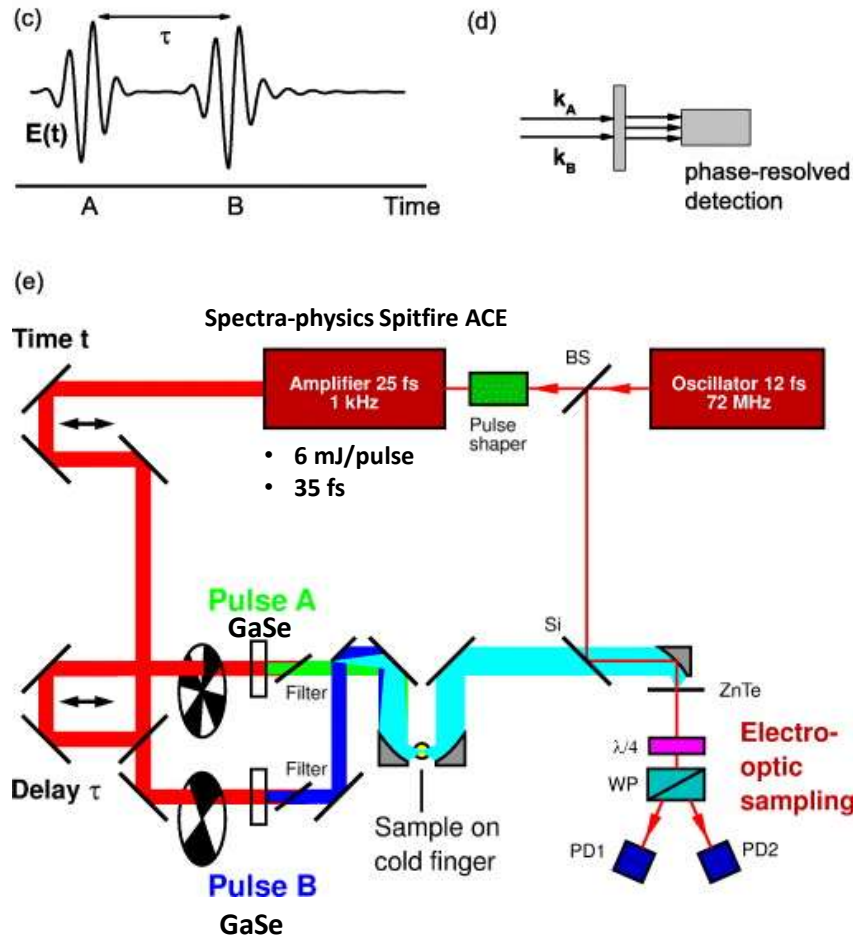
Phys. Rev. Lett. **124**, 117205 (2020)

Two types of spectral broadening:

- 1) Inhomogeneous broadening- results from a difference between members on an ensemble
- 2) Homogenous broadening-intrinsic lifetime broadening

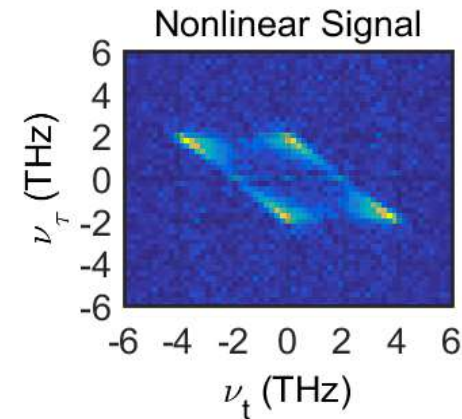
2DCS can distinguish between these two sources of broadening by spreading the spectrum in the frequency plane

2D Coherent Spectroscopy at LUMOS



- Collinear geometry with phase stabilized THz pulses
- Frequency plane defined by ν_t = bandwidth of THz and ν_τ = time delay between pulses
- Nonlinear 2DCS signal extracted from the linear response

$$E_{NL} = E_{A+B} - E_A - E_B$$



Bi₂Se₃ data Courtesy of P. Bowlan (C-PCS)

Key Technical Challenges: Enhancing THz Field Strength and S/N

2DCS is an inherently nonlinear ($\chi^{(3)}$) process: Requires high THz field strengths over a considerably large diameter of the THz beam/focal spot as well as good S/N

Three approaches:

- 1) Enhance THz field strength @ generation. Ex: tilted pulse front, Organic xtal
- 2) Employ high repetition rate lasers
- 3) Use single shot EOS detection

(Minor) Setbacks

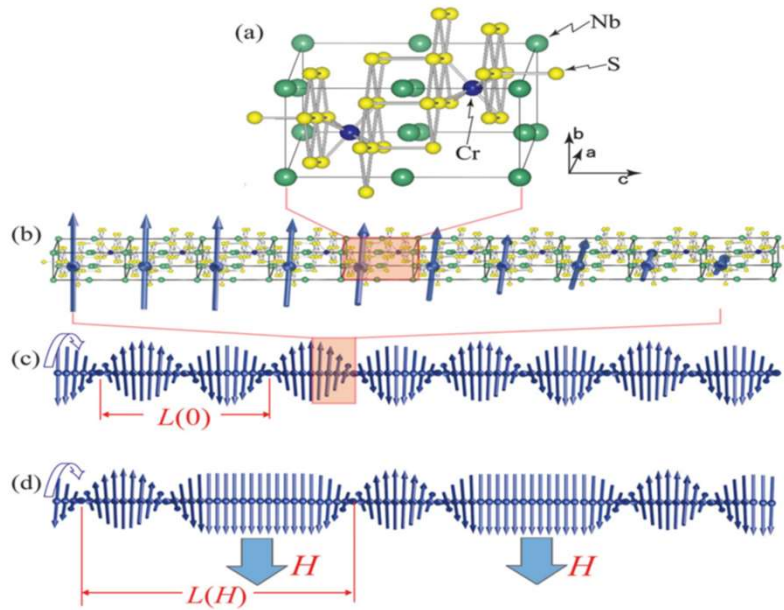
Technical

- Difficulty in sourcing samples
Continue to foster collaboration with both internal and external sample growers
- Insufficient gain from our 6 mJ, Ti:Sapphire regenerative amplifier due to damaged optics in the cavity
Service visit to repair laser system is scheduled with work resuming later this month

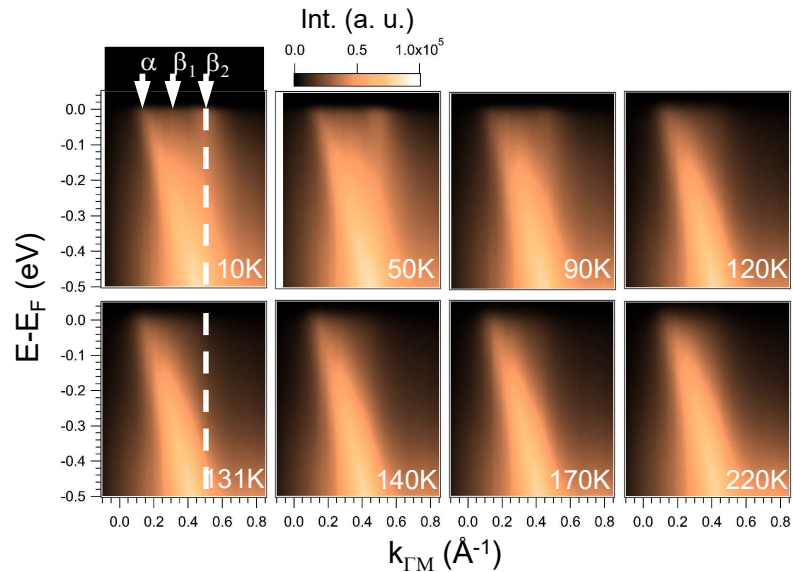
Personnel

- Postdoc hired prior to start date of project but arrival to LANL delayed due to (partially COVID related) issues with H1B

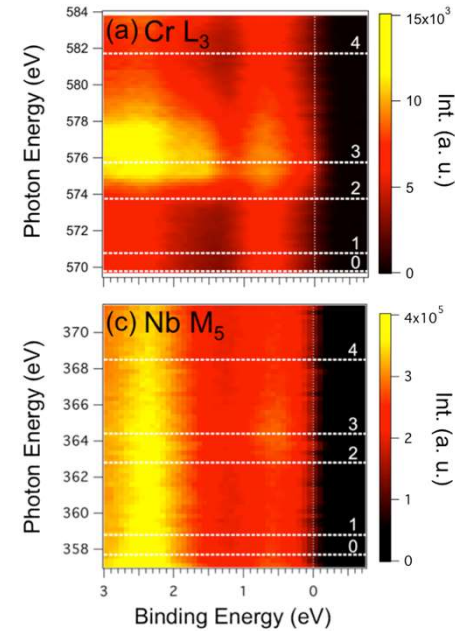
Disentangling the Electronic, Lattice, and Spin Dynamics in $\text{Cr}_{1/3}\text{NbS}_2$



Phys. Rev. Lett. **108**, 107202 (2012)

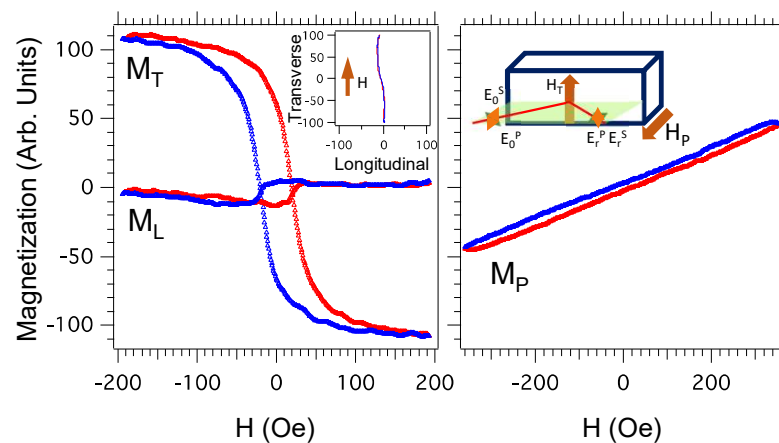


Comm. Phys. **3**, 65 (2020)

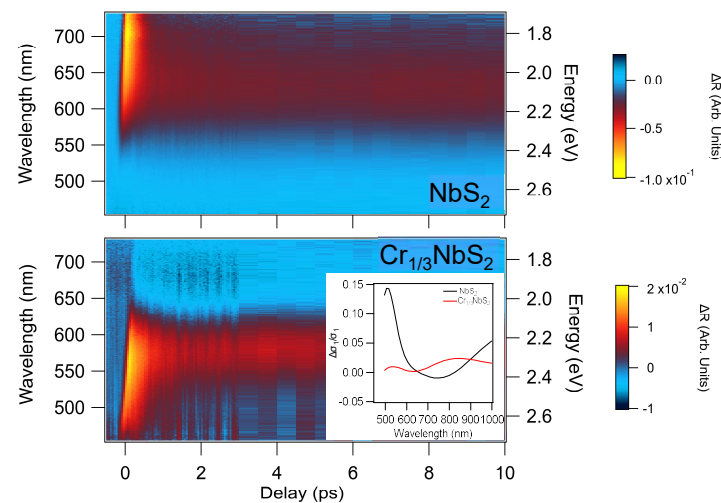


- Anisotropic chiral helimagnet $\text{Cr}_{1/3}\text{NbS}_2$: Application of B-field perpendicular to helical axis induces a soliton lattice phase
- Significant effect on electronic transport: Electronic structure found to play a non-trivial role
- Can this CSL phase be tuned with light? How does shifting spectral weight and presence of hybridized Cr/Nb states near EF influence the dynamics?

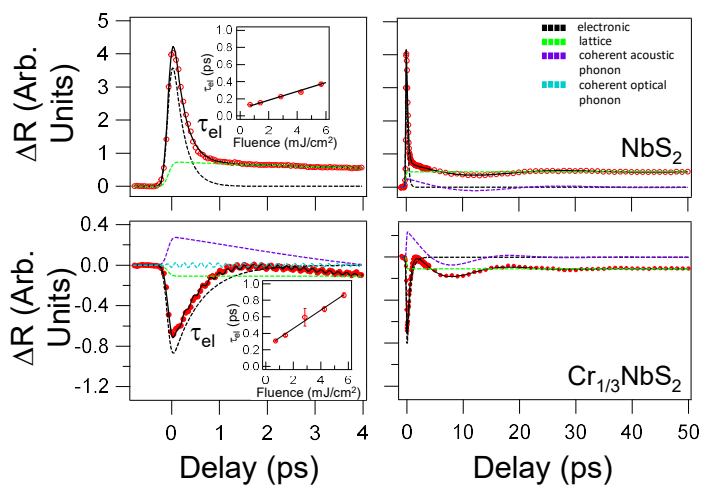
Static MOKE



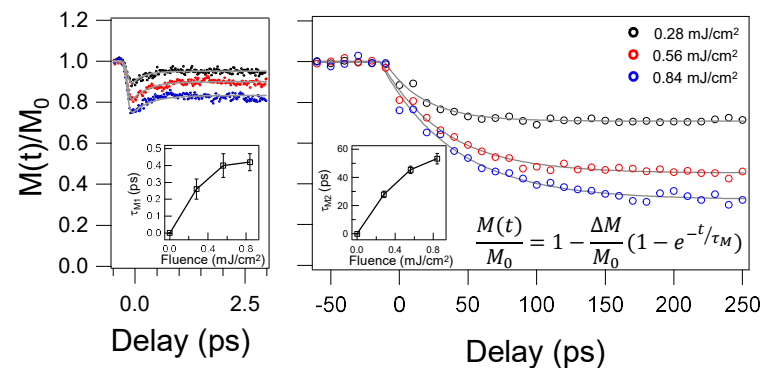
Broadband Probe



Transient Reflectivity



Time-Resolved MOKE



Acknowledgements

Resolving the Spinon Continuum:

M.-C. Lee, Y. Huang, and R. P. Prasankumar

[MPA-CINT](#)

P. Bowlan

[C-PCS](#)

S. Lin

[T-4](#)

Disentangling the Electronic, Lattice and Spin Dynamics of $\text{Cr}_{1/3}\text{NbS}_2$

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